

03/25/02

03-26-02

1995-03-25 MAR 25 2002

FORM PCT-1390
(REV. 01-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

1576.100

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

10/089131

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/JP00/06892

04 October 2000 (4.10.00)

04 October 1999 (4.10.99)

TITLE OF INVENTION PHENOL COMPOUNDS AND RECORDING MATERIALS USING THE SAME

APPLICANT(S) FOR DO/EO/US HIDAKA ET AL., Tomoya

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☐ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

Copy of Form PCT/IB/304; Copy of International Search Report; Copy of PCT Application (Japanese); Copy of Preliminary Examination (Japanese); Copy of WO 01/21593 AI (Japanese)

U.S. APPLICATION NO. of known case 37 CFR 1.5 1070891 31		INTERNATIONAL APPLICATION NO PCT/JP00/06892		ATTORNEY'S DOCKET NUMBER 1576.100	
--------------------------------------------------------------------	--	-------------------------------------------------------	--	---------------------------------------------	--

21. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a) (2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT =				CALCULATIONS PTO USE ONLY	
				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 0.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	6 -20 =	0	x \$18.00	\$ 0.00	
Independent claims	6 -3 =	3	x \$84.00	\$ 252.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				\$ 0.00	
TOTAL OF ABOVE CALCULATIONS =				\$ 1,142.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$ 0.00	
SUBTOTAL =				\$ 1,142.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$ 0.00	
TOTAL FEES ENCLOSED =				\$ 1,142.00	
				Amount to be refunded:	\$
				charged:	\$

a. ☐ A check in the amount of \$ _____ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 131992 in the amount of \$ 1,142.00 to cover the above fees.
 A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
 overpayment to Deposit Account No. 131992. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card
 information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Dennis G. LaPointe
 Mason & Associates, P.A.
 17757 US Hwy 19 N., Suite 500
 Clearwater, FL 33764

Dennis G. LaPointe
SIGNATURE

Dennis G. LaPointe
NAME

40,693
REGISTRATION NUMBER

PHENOL COMPOUNDS AND RECORDING MATERIALS USING THE SAME

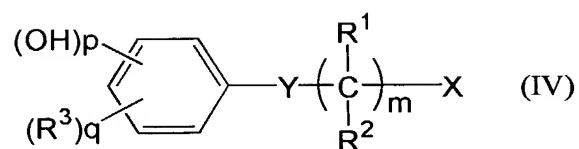
Technical Field of Invention

The present invention is related to novel phenol compounds and recording materials containing the phenol compound and having excellent image storing and stabilizing properties.

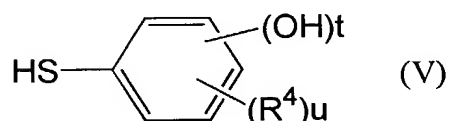
Background Art

Recording materials employing a manner of coloring by a reaction of a color forming dye and a developer have been widely used in thermal recording papers for recording outputted information from facsimiles, printers, etc. and pressure-sensitive copying papers for concurrently producing a plurality of copies, because such recording materials enable to record images in a short time by employing a relatively simple apparatus without requiring complex process such as development and fixation. As such recording materials, a material capable of instantly color forming, keeping whiteness of the part where no color is formed, hereinafter referred to as "background", and providing high hardness of the color formed images is required. However, in view of stability during storing in long-term basis, a recording material capable of providing excellent lightfastness to the color formed images is particularly desired. In this concern, development of color forming dyes, developers, stabilizers during storing, etc. has been tried in the field of this industry, however, a material having excellent sensitivity in color forming, giving whiteness on the background and image stability with a good balance and an enough satisfaction has not been obtained.

As compounds that are related to the present invention, in Jap. Pat. Appln. KOKAI Publication Nos. 2-204091, 1-72891 and 4-217657, the phenol compounds are disclosed as examples for a developer. In these disclosures, however, a technique to provide a recording material having high performance in the background effect and image stabilizing effects is sought. In addition, compounds similar to the compounds of the present invention are



Wherein R^1 , R^2 , R^3 , Y , m , p and q are as defined above, X represents halogen, such as chlorine and bromine, to a reaction with a compound represented by a general formula (V);

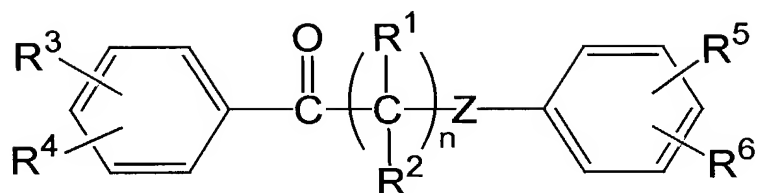


wherein R^4 , t and u are as defined above, in an organic solvent, for example, methanol in the presence of a base.

Compounds represented by the general formula (I), wherein the portion of S(O)_n is SO or SO_2 , may be obtained by oxidizing the compound obtained by the reaction hereinabove with an oxidizing agent, such as aqueous solution of hydrogen peroxide and *m*-chloroperbenzoic acid in an appropriate solvent.

The compounds those, which may be synthesized according to the process, described above are presented in Tables 1 and 2.

Table 1



Compound No.	R ³	R ⁴	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point (°C)
I-1	2-OH	H	H	H	1	S	H	H	
I-2	2-OH	H	H	H	1	SO	H	H	
I-3	2-OH	H	H	H	1	SO ₂	H	H	
I-4	2-OH	H	H	H	1	S	4-OH	H	139-141
I-5	2-OH	H	H	H	1	SO	4-OH	H	166-167
I-6	2-OH	H	H	H	1	SO ₂	4-OH	H	143-146
I-7	2-OH	H	H	H	2	S	4-OH	H	
I-8	2-OH	H	H	H	2	SO	4-OH	H	
I-9	2-OH	H	H	H	2	SO ₂	4-OH	H	
I-10	2-OH	H	H	H	3	S	4-OH	H	
I-11	2-OH	H	H	H	3	SO	4-OH	H	
I-12	2-OH	H	H	H	3	SO ₂	4-OH	H	
I-13	2-OH	H	H	H	4	S	4-OH	H	
I-14	2-OH	H	H	H	4	SO	4-OH	H	
I-15	2-OH	H	H	H	4	SO ₂	4-OH	H	
I-16	2-OH	5-CH ₃	H	H	1	S	4-OH	H	
I-17	2-OH	5-CH ₃	H	H	1	SO	4-OH	H	
I-18	2-OH	5-CH ₃	H	H	1	SO ₂	4-OH	H	
I-19	2-OH	5-Cl	H	H	1	S	4-OH	H	
I-20	2-OH	5-Cl	H	H	1	SO	4-OH	H	
I-21	2-OH	5-Cl	H	H	1	SO ₂	4-OH	H	
I-22	2-OH	5-Br	H	H	1	S	4-OH	H	
I-23	2-OH	5-Br	H	H	1	SO	4-OH	H	
I-24	2-OH	5-Br	H	H	1	SO ₂	4-OH	H	
I-25	2-OH	4-OCH ₃	H	H	1	S	4-OH	H	
I-26	2-OH	4-OCH ₃	H	H	1	SO	4-OH	H	
I-27	2-OH	4-OCH ₃	H	H	1	SO ₂	4-OH	H	
I-28	2-OH	5-OCH ₃	H	H	1	S	4-OH	H	
I-29	2-OH	5-OCH ₃	H	H	1	SO	4-OH	H	
I-30	2-OH	5-OCH ₃	H	H	1	SO ₂	4-OH	H	

Table 1 (Continued)

Compound No.	R ³	R ⁴	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point (°C)
I-31	2-OH	H	CH ₃	H	1	S	4-OH	H	
I-32	2-OH	H	CH ₃	H	1	SO	4-OH	H	
I-33	2-OH	H	CH ₃	H	1	SO ₂	4-OH	H	
I-34	2-OH	H	CH ₃	CH ₃	1	S	4-OH	H	
I-35	2-OH	H	CH ₃	CH ₃	1	SO	4-OH	H	
I-36	2-OH	H	CH ₃	CH ₃	1	SO ₂	4-OH	H	
I-37	2-OH	H	H	H	1	S	2-OH	4-OH	
I-38	2-OH	H	H	H	1	SO	2-OH	4-OH	
I-39	2-OH	H	H	H	1	SO ₂	2-OH	4-OH	
I-40	2-OH	H	H	H	1	S	2-OH	5-OH	
I-41	2-OH	H	H	H	1	SO	2-OH	5-OH	
I-42	2-OH	H	H	H	1	SO ₂	2-OH	5-OH	
I-43	2-OH	H	H	H	1	S	2-OH	5-CH ₃	
I-44	2-OH	H	H	H	1	SO	2-OH	5-CH ₃	
I-45	2-OH	H	H	H	1	SO ₂	2-OH	5-CH ₃	
I-46	2-OH	H	H	H	1	S	3-CH ₃	4-OH	
I-47	2-OH	H	H	H	1	SO	3-CH ₃	4-OH	
I-48	2-OH	H	H	H	1	SO ₂	3-CH ₃	4-OH	
I-49	2-OH	H	H	H	1	S	3-Cl	4-OH	
I-50	2-OH	H	H	H	1	SO	3-Cl	4-OH	
I-51	2-OH	H	H	H	1	SO ₂	3-Cl	4-OH	
I-52	2-OH	H	H	H	1	S	2-CH ₃	4-OH	
I-53	2-OH	H	H	H	1	SO	2-CH ₃	4-OH	
I-54	2-OH	H	H	H	1	SO ₂	2-CH ₃	4-OH	
I-55	3-OH	H	H	H	1	S	H	H	
I-56	3-OH	H	H	H	1	SO	H	H	
I-57	3-OH	H	H	H	1	SO ₂	H	H	
I-58	3-OH	H	H	H	1	S	4-OH	H	156-159
I-59	3-OH	H	H	H	1	SO	4-OH	H	155-157
I-60	3-OH	H	H	H	1	SO ₂	4-OH	H	189-192
I-61	3-OH	H	H	H	2	S	4-OH	H	
I-62	3-OH	H	H	H	2	SO	4-OH	H	
I-63	3-OH	H	H	H	2	SO ₂	4-OH	H	
I-64	3-OH	H	H	H	3	S	4-OH	H	
I-65	3-OH	H	H	H	3	SO	4-OH	H	
I-66	3-OH	H	H	H	3	SO ₂	4-OH	H	
I-67	3-OH	H	H	H	4	S	4-OH	H	
I-68	3-OH	H	H	H	4	SO	4-OH	H	
I-69	3-OH	H	H	H	4	SO ₂	4-OH	H	

Table 1 (Continued)

Compound No.	R ³	R ⁴	R ¹	R ²	n	Z	R ⁵	R ^b	Melting Point (°C)
I-70	3-OH	5-CH ₃	H	H	1	S	4-OH	H	
I-71	3-OH	5-CH ₃	H	H	1	SO	4-OH	H	
I-72	3-OH	5-CH ₃	H	H	1	SO ₂	4-OH	H	
I-73	3-OH	5-Cl	H	H	1	S	4-OH	H	
I-74	3-OH	5-Cl	H	H	1	SO	4-OH	H	
I-75	3-OH	5-Cl	H	H	1	SO ₂	4-OH	H	
I-76	3-OH	4-OCCH ₃	H	H	1	S	4-OH	H	
I-77	3-OH	4-OCCH ₃	H	H	1	SO	4-OH	H	
I-78	3-OH	4-OCCH ₃	H	H	1	SO ₂	4-OH	H	
I-79	3-OH	H	CH ₃	H	1	S	4-OH	H	
I-80	3-OH	H	CH ₃	H	1	SO	4-OH	H	
I-81	3-OH	H	CH ₃	H	1	SO ₂	4-OH	H	
I-82	3-OH	H	CH ₃	CH ₃	1	S	4-OH	H	
I-83	3-OH	H	CH ₃	CH ₃	1	SO	4-OH	H	
I-84	3-OH	H	CH ₃	CH ₃	1	SO ₂	4-OH	H	
I-85	3-OH	H	H	H	1	S	2-OH	4-OH	
I-86	3-OH	H	H	H	1	SO	2-OH	4-OH	
I-87	3-OH	H	H	H	1	SO ₂	2-OH	4-OH	
I-88	3-OH	H	H	H	1	S	2-OH	5-OH	
I-89	3-OH	H	H	H	1	SO	2-OH	5-OH	
I-90	3-OH	H	H	H	1	SO ₂	2-OH	5-OH	
I-91	3-OH	H	H	H	1	S	2-OH	5-CH ₃	
I-92	3-OH	H	H	H	1	SO	2-OH	5-CH ₃	
I-93	3-OH	H	H	H	1	SO ₂	2-OH	5-CH ₃	
I-94	3-OH	H	H	H	1	S	3-CH ₃	4-OH	
I-95	3-OH	H	H	H	1	SO	3-CH ₃	4-OH	
I-96	3-OH	H	H	H	1	SO ₂	3-CH ₃	4-OH	
I-97	3-OH	H	H	H	1	S	3-Cl	4-OH	
I-98	3-OH	H	H	H	1	SO	3-Cl	4-OH	
I-99	3-OH	H	H	H	1	SO ₂	3-Cl	4-OH	
I-100	3-OH	H	H	H	1	S	2-CH ₃	4-OH	
I-101	3-OH	H	H	H	1	SO	2-CH ₃	4-OH	
I-102	3-OH	H	H	H	1	SO ₂	2-CH ₃	4-OH	
I-103	4-OH	H	H	H	1	S	H	H	168-171
I-104	4-OH	H	H	H	1	SO	H	H	
I-105	4-OH	H	H	H	1	SO ₂	H	H	154-156
I-106	4-OH	H	H	H	1	S	4-OH	H	194-197
I-107	4-OH	H	H	H	1	SO	4-OH	H	167-169
I-108	4-OH	H	H	H	1	SO ₂	4-OH	H	212-214

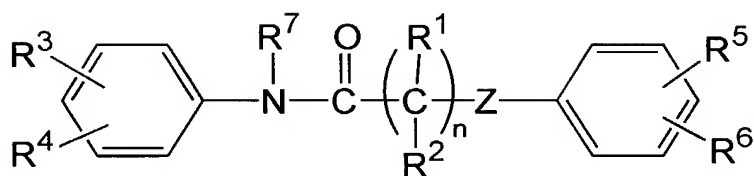
Table 1 (Continued)

Compound No.	R ³	R ⁴	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point (°C)
I-109	4-OH	H	H	H	2	S	4-OH	H	
I-110	4-OH	H	H	H	2	SO	4-OH	H	
I-111	4-OH	H	H	H	2	SO ₂	4-OH	H	
I-112	4-OH	H	H	H	3	S	4-OH	H	
I-113	4-OH	H	H	H	3	SO	4-OH	H	
I-114	4-OH	H	H	H	3	SO ₂	4-OH	H	
I-115	4-OH	H	H	H	4	S	4-OH	H	
I-116	4-OH	H	H	H	4	SO	4-OH	H	
I-117	4-OH	H	H	H	4	SO ₂	4-OH	H	
I-118	2-CH ₃	4-OH	H	H	1	S	4-OH	H	94-96
I-119	2-CH ₃	4-OH	H	H	1	SO	4-OH	H	
I-120	2-CH ₃	4-OH	H	H	1	SO ₂	4-OH	H	187-189
I-121	3-CH ₃	4-OH	H	H	1	S	4-OH	H	
I-122	3-CH ₃	4-OH	H	H	1	SO	4-OH	H	
I-123	3-CH ₃	4-OH	H	H	1	SO ₂	4-OH	H	
I-124	3-Cl	4-OH	H	H	1	S	4-OH	H	
I-125	3-Cl	4-OH	H	H	1	SO	4-OH	H	
I-126	3-Cl	4-OH	H	H	1	SO ₂	4-OH	H	
I-127	3-Br	4-OH	H	H	1	S	4-OH	H	
I-128	3-Br	4-OH	H	H	1	SO	4-OH	H	
I-129	3-Br	4-OH	H	H	1	SO ₂	4-OH	H	
I-130	3-CH ₃	4-OH	H	H	1	S	4-OH	H	
I-131	3-CH ₃	4-OH	H	H	1	SO	4-OH	H	
I-132	3-CH ₃	4-OH	H	H	1	SO ₂	4-OH	H	
I-133	4-OH	H	CH ₃	H	1	S	4-OH	H	
I-134	4-OH	H	CH ₃	H	1	SO	4-OH	H	
I-135	4-OH	H	CH ₃	H	1	SO ₂	4-OH	H	
I-136	4-OH	H	CH ₃	CH ₃	1	S	4-OH	H	
I-137	4-OH	H	CH ₃	CH ₃	1	SO	4-OH	H	
I-138	4-OH	H	CH ₃	CH ₃	1	SO ₂	4-OH	H	
I-139	4-OH	H	H	H	1	S	2-OH	4-OH	178-180
I-140	4-OH	H	H	H	1	SO	2-OH	4-OH	
I-141	4-OH	H	H	H	1	SO ₂	2-OH	4-OH	224-226
I-142	4-OH	H	H	H	1	S	2-OH	5-OH	
I-143	4-OH	H	H	H	1	SO	2-OH	5-OH	
I-144	4-OH	H	H	H	1	SO ₂	2-OH	5-OH	
I-145	4-OH	H	H	H	1	S	2-OH	5-CH ₃	145-147
I-146	4-OH	H	H	H	1	SO	2-OH	5-CH ₃	
I-147	4-OH	H	H	H	1	SO ₂	2-OH	5-CH ₃	180-183

Table 1 (Continued)

Compound No.	R ³	R ⁴	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point (°C)
I-148	4-OH	H	H	H	1	S	3-CH ₃	4-OH	
I-149	4-OH	H	H	H	1	SO	3-CH ₃	4-OH	
I-150	4-OH	H	H	H	1	SO ₂	3-CH ₃	4-OH	
I-151	4-OH	H	H	H	1	S	3-Cl	4-OH	
I-152	4-OH	H	H	H	1	SO	3-Cl	4-OH	
I-153	4-OH	H	H	H	1	SO ₂	3-Cl	4-OH	
I-154	4-OH	H	H	H	1	S	2-CH ₃	4-OH	150-152
I-155	4-OH	H	H	H	1	SO	2-CH ₃	4-OH	
I-156	4-OH	H	H	H	1	SO ₂	2-CH ₃	4-OH	207-209
I-157	4-NO ₂	H	H	H	1	SO ₂	4-OH	H	184-186
I-158	4-OH	2-OH	H	H	1	S	4-OH	H	122-125

Table 2



Compound No.	R ³	R ⁴	R ⁷	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point
II-1	H	H	H	H	H	1	SO	4-OH	H	208-210
II-2	H	H	H	H	H	1	SO ₂	4-OH	H	188-189
II-3	H	H	H	H	H	2	SO	4-OH	H	
II-4	H	H	H	H	H	2	SO ₂	4-OH	H	191-193
II-5	H	H	H	H	H	1	SO	2-OH	4-OH	
II-6	H	H	H	H	H	1	SO ₂	2-OH	4-OH	222-224
II-7	H	H	H	H	H	1	SO	2-OH	5-OH	
II-8	H	H	H	H	H	1	SO ₂	2-OH	5-OH	
II-9	H	H	H	H	H	1	SO	2-CH ₃	4-OH	
II-10	H	H	H	H	H	1	SO ₂	2-CH ₃	4-OH	
II-11	H	H	H	H	H	1	SO	2-CH ₃	4-OH	
II-12	H	H	H	H	H	1	SO ₂	2-CH ₃	4-OH	
II-13	H	H	H	H	H	1	SO	2-CH ₃	5-OH	
II-14	H	H	H	H	H	1	SO ₂	2-CH ₃	5-OH	
II-15	H	H	CH ₃	H	H	1	SO	4-OH	H	138-139
II-16	H	H	CH ₃	H	H	1	SO ₂	4-OH	H	194-196
II-17	2-CH ₃	H	H	H	H	1	SO	4-OH	H	
II-18	2-CH ₃	H	H	H	H	1	SO ₂	4-OH	H	
II-19	3-CH ₃	H	H	H	H	1	SO	4-OH	H	
II-20	3-CH ₃	H	H	H	H	1	SO ₂	4-OH	H	
II-21	4-CH ₃	H	H	H	H	1	SO	4-OH	H	
II-22	4-CH ₃	H	H	H	H	1	SO ₂	4-OH	H	203-204
II-23	4-Cl	H	H	H	H	1	SO	4-OH	H	
II-24	4-Cl	H	H	H	H	1	SO ₂	4-OH	H	212-213
II-25	4-Br	H	H	H	H	1	SO	4-OH	H	
II-26	4-Br	H	H	H	H	1	SO ₂	4-OH	H	
II-27	2-OCH ₃	H	H	H	H	1	SO	4-OH	H	
II-28	2-OCH ₃	H	H	H	H	1	SO ₂	4-OH	H	158-161
II-29	3-OCH ₃	H	H	H	H	1	SO	4-OH	H	
II-30	3-OCH ₃	H	H	H	H	1	SO ₂	4-OH	H	178-180
II-31	4-OCH ₃	H	H	H	H	1	SO	4-OH	H	
II-32	4-OCH ₃	H	H	H	H	1	SO ₂	4-OH	H	185-188

Table 2 (Continued)

Compound No.	R ³	R ⁴	R ⁷	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point
II-33	2-CO ₂ CH ₃	H	H	H	H	1	S0	4-OH	H	
II-34	2-CO ₂ CH ₃	H	H	H	H	1	S0 ₂	4-OH	H	
II-35	3-CO ₂ CH ₃	H	H	H	H	1	S0	4-OH	H	
II-36	3-CO ₂ CH ₃	H	H	H	H	1	S0 ₂	4-OH	H	
II-37	4-CO ₂ CH ₃	H	H	H	H	1	S0	4-OH	H	
II-38	4-CO ₂ CH ₃	H	H	H	H	1	S0 ₂	4-OH	H	232-235
II-39	3-CO ₂ CH ₂ CH ₃	H	H	H	H	1	S0	4-OH	H	
II-40	3-CO ₂ CH ₂ CH ₃	H	H	H	H	1	S0 ₂	4-OH	H	
II-41	4-CO ₂ CH ₂ CH ₃	H	H	H	H	1	S0	4-OH	H	
II-42	4-CO ₂ CH ₂ CH ₃	H	H	H	H	1	S0 ₂	4-OH	H	203-205
II-43	2-CO ₂ H	H	H	H	H	1	S0	4-OH	H	
II-44	2-CO ₂ H	H	H	H	H	1	S0 ₂	4-OH	H	
II-45	3-CO ₂ H	H	H	H	H	1	S0	4-OH	H	
II-46	3-CO ₂ H	H	H	H	H	1	S0 ₂	4-OH	H	
II-47	4-CO ₂ H	H	H	H	H	1	S0	4-OH	H	
II-48	4-CO ₂ H	H	H	H	H	1	S0 ₂	4-OH	H	285-286
II-49	3-CONHCH ₃	H	H	H	H	1	S0	4-OH	H	
II-50	3-CONHCH ₃	H	H	H	H	1	S0 ₂	4-OH	H	
II-51	4-CONHPh	H	H	H	H	1	S0	4-OH	H	
II-52	4-CONHPh	H	H	H	H	1	S0 ₂	4-OH	H	
II-53	3-CON(CH ₃) ₂	H	H	H	H	1	S0	4-OH	H	
II-54	3-CON(CH ₃) ₂	H	H	H	H	1	S0 ₂	4-OH	H	
II-55	2-SO ₂ NH ₂	H	H	H	H	1	S0	4-OH	H	
II-56	2-SO ₂ NH ₂	H	H	H	H	1	S0 ₂	4-OH	H	
II-57	3-SO ₂ NH ₂	H	H	H	H	1	S0	4-OH	H	
II-58	3-SO ₂ NH ₂	H	H	H	H	1	S0 ₂	4-OH	H	
II-59	4-SO ₂ NH ₂	H	H	H	H	1	S0	4-OH	H	
II-60	4-SO ₂ NH ₂	H	H	H	H	1	S0 ₂	4-OH	H	285-287
II-61	4-SO ₂ NHPh	H	H	H	H	1	S0	4-OH	H	
II-62	4-SO ₂ NHPh	H	H	H	H	1	S0 ₂	4-OH	H	
II-63	H	H	Ph	H	H	1	S0	4-OH	H	
II-64	H	H	Ph	H	H	1	S0 ₂	4-OH	H	265-269
II-65	2-OH	H	H	H	H	1	S	4-OH	H	176-179
II-66	2-OH	H	H	H	H	1	S0	4-OH	H	
II-67	2-OH	H	H	H	H	1	S0 ₂	4-OH	H	190-192
II-68	2-OH	4-CH ₃	H	H	H	1	S	4-OH	H	
II-69	2-OH	4-CH ₃	H	H	H	1	S0	4-OH	H	
II-70	2-OH	4-CH ₃	H	H	H	1	S0 ₂	4-OH	H	

Table 2 (Continued)

Compound No.	R ³	R ⁴	R ⁷	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point
II-71	2-OH	4-OCH ₃	H	H	H	1	S	4-OH	H	
II-72	2-OH	4-OCH ₃	H	H	H	1	SO	4-OH	H	
II-73	2-OH	4-OCH ₃	H	H	H	1	SO ₂	4-OH	H	
II-74	2-OH	H	H	H	H	1	S	H	H	141-142
II-75	2-OH	H	H	H	H	1	SO	H	H	
II-76	2-OH	H	H	H	H	1	SO ₂	H	H	
II-77	2-OH	H	CH ₃	H	H	1	S	4-OH	H	
II-78	2-OH	H	CH ₃	H	H	1	SO	4-OH	H	
II-79	2-OH	H	CH ₃	H	H	1	SO ₂	4-OH	H	
II-80	2-OH	H	Ph	H	H	1	S	4-OH	H	
II-81	2-OH	H	Ph	H	H	1	SO	4-OH	H	
II-82	2-OH	H	Ph	H	H	1	SO ₂	4-OH	H	
II-83	3-OH	H	H	H	H	1	S	4-OH	H	171-173
II-84	3-OH	H	H	H	H	1	SO	4-OH	H	202-204
II-85	3-OH	H	H	H	H	1	SO ₂	4-OH	H	254-256
II-86	3-OH	4-CH ₃	H	H	H	1	S	4-OH	H	181-182
II-87	3-OH	4-CH ₃	H	H	H	1	SO	4-OH	H	
II-88	3-OH	4-CH ₃	H	H	H	1	SO ₂	4-OH	H	200-203
II-89	3-OH	H	H	H	H	1	S	H	H	
II-90	3-OH	H	H	H	H	1	SO	H	H	
II-91	3-OH	H	H	H	H	1	SO ₂	H	H	
II-92	3-OH	H	CH ₃	H	H	1	S	4-OH	H	
II-93	3-OH	H	CH ₃	H	H	1	SO	4-OH	H	
II-94	3-OH	H	CH ₃	H	H	1	SO ₂	4-OH	H	
II-95	3-OH	H	Ph	H	H	1	S	4-OH	H	158-159
II-96	3-OH	H	Ph	H	H	1	SO	4-OH	H	191-192
II-97	3-OH	H	Ph	H	H	1	SO ₂	4-OH	H	238-239
II-98	4-OH	H	H	H	H	1	S	4-OH	H	163-164
II-99	4-OH	H	H	H	H	1	SO	4-OH	H	220-221
II-100	4-OH	H	H	H	H	1	SO ₂	4-OH	H	211-215
II-101	4-OH	H	H	H	H	1	S	H	H	
II-102	4-OH	H	H	H	H	1	SO	H	H	
II-103	4-OH	H	H	H	H	1	SO ₂	H	H	
II-104	4-OH	H	CH ₃	H	H	1	S	4-OH	H	
II-105	4-OH	H	CH ₃	H	H	1	SO	4-OH	H	
II-106	4-OH	H	CH ₃	H	H	1	SO ₂	4-OH	H	
II-107	4-OH	H	Ph	H	H	1	S	4-OH	H	180-181
II-108	4-OH	H	Ph	H	H	1	SO	4-OH	H	215-217
II-109	4-OH	H	Ph	H	H	1	SO ₂	4-OH	H	276-277

Table 2 (Continued)

Compound No.	R ³	R ⁴	R ⁷	R ¹	R ²	n	Z	R ⁵	R ⁶	Melting Point
II-110	3-OH	H	(3-OH)Ph	H	H	1	S	4-OH	H	
II-111	3-OH	H	(3-OH)Ph	H	H	1	SO	4-OH	H	
II-112	3-OH	H	(3-OH)Ph	H	H	1	SO ₂	4-OH	H	
II-113	4-OH	H	(4-OH)Ph	H	H	1	S	4-OH	H	
II-114	4-OH	H	(4-OH)Ph	H	H	1	SO	4-OH	H	
II-115	4-OH	H	(4-OH)Ph	H	H	1	SO ₂	4-OH	H	
II-116	2-CH ₃	4-OCH ₃	H	H	H	1	SO ₂	4-OH	H	164-167
II-117	4-SO ₂ NH ₂	2-OH	H	H	H	1	S	4-OH	H	221-225
II-118	3-OCH ₃	H	Ph	H	H	1	SO ₂	4-OH	H	205-208
II-119	4-OCH ₃	2-CH ₃	Ph	H	H	1	SO ₂	4-OH	H	228-230
II-120	3-OH	H	c-Hexyl	H	H	1	S	4-OH	H	193-196
II-121	3-OH	H	c-Hexyl	H	H	1	SO ₂	4-OH	H	240-243
II-122	2-OH	H	H	H	H	2	S	4-OH	H	134-139
II-123	2-OH	H	H	H	H	2	SO ₂	4-OH	H	156-157
II-124	2-NO ₂	4-OCH ₃	H	H	H	2	SO ₂	4-OH	H	130-132
II-125	2-OH	H	H	CH ₃	H	1	S	4-OH	H	166-171
II-126	2-OH	4-NO ₂	H	H	H	1	S	4-OH	H	232-233
II-127	2-OH	5-Cl	H	H	H	1	S	4-OH	H	185-186
II-128	2-OH	5-CH ₃	H	H	H	1	S	4-OH	H	174-176

The present invention may be applied for any use for recording materials as far as they employ a color forming dye, for example, for thermal recording materials and pressure-sensitive copying materials.

When the present invention is used for thermal recording papers, it can be applied according to the same method for applying a known stabilizer for image keeping and developer. For example, each of a compound of the present invention in fine particles and a color forming dye in fine particles are dispersed in an aqueous solution of an aqueous binding agent, such as polyvinyl alcohol and cellulose, and the resultant suspension is mixed and coated onto a support material, for example a paper, and then dried to obtain a thermal recording paper.

The ratio of the compound represented by the general formula (I) of the present invention to be used with respect to a color forming dye is 1 to 10 parts by weight based on 1 part by weight

of a color forming dye, and preferably 1.5 to 5 parts by weight.

The recording material of the present invention may also contain one or more of a known developer, an image stabilizer, a sensitizer, a filler, a dispersing agent, an antioxidant, a desensitizer, an antitack agent, an antifoamer, a photo stabilizer, a fluorescence brightener, and the like upon requirement in addition to a color forming dye and the compound represented by the general formula (I).

Examples of the color forming dye used for the recording material of the present invention include leuco dyes based on fluoran, phthalide, lactam, triphenyl methane, fenothiazine, and spiropyran. However, the color forming dyes are not limited to these leuco dyes, and any color forming dyes may be used as far as it forms color by contacting with a developer of an acidic substance. Each of these color forming dyes can form a color independently, and it naturally constitutes a recording material having a color that is formed by the color forming dye, and two or more of these color forming dyes may be used in combination. For example, a recording material that forms true black may be prepared by combining color forming dyes each forming red, blue and green and a color forming dye forming black.

anilinofluoran, 3-diethylamino-7-(o-chloroanilino)fluoran, 3-dibutylamino-7-(o-chloroanilino)fluoran, 3-(N-ethyl-p-toluidino)-6-methyl-7-anilinofluoran, 3-(N-cyclohexyl-N-methylamino)-6-methyl-7-anilinofluoran, 3-pyrrolidino-6-methyl-7-aralinofluoran, 3-piperidino-6-methyl-7-aralinofluoran, 3-dimethylamino-7-(m-trifluoromethylanilino)fluoran, 3-dipentylamino-6-methyl-7-anilinofluoran, 3-(N-ethoxypropyl-N-ethylamino)-6-methyl-7-anilinofluoran, 3-dibutylamino-7-(o-fluoroanilino)fluoran, 3-diethylaminobenzo[a]fluoran, 3-dimethylamino-6-methyl-7-chlorofluoran, 3-diethylamino-5-methyl-7-dibenzylaminofluoran, 3-diethylamino-7-dibenzylaminofluoran, 3-diethylamino-5-chlorofluoran, 3-diethylamino-6-(N,N'-dibenzylamino)fluoran, 3,6-domethoxyfluoran, and 2,4-dimethyl-6-(4-dimethylaminophenyl)aminofluoran.

As a near infrared absorbing dye, 3-(4-(4-(4-anilino)-anilino)anilino-6-methyl-7-chlorofluoran, 3,3-bis(2-(4-dimethylaminophenyl)-2-(4-methoxyphenyl)vinyl)-4,5,6,7-tetrachlorophthalide, 3,6,6'-tris(dimethylamino)spiro[fluorine-9,3'-phthalide and the like may be exemplified.

Other than the above, 3,3-bis(4'-diethylaminophenyl)-6-diethylaminophthalide and the like are also exemplified.

Examples for the developer include bisphenol compounds, such as bisphenol A, 4,4'-sec-butyldienebisphenol, 4,4'-cyclohexylidenebisphenol, 2,2-dimethyl-3,3-bis(4-hydroxyphenyl)butane, 2,2'-dihydroxydiphenyl, pentamethylene-bis(4-hydroxybenzoate), 2,2-dimethyl-3,3-di(4-hydroxyphenyl)pentane, 2,2-di(4-hydroxyphenyl)hexane; metal salts of benzoic acid, such as zinc benzoate and zinc 4-nitrobenzoate; salicylates, such as salicylic 4-(2-(4-methoxyphenyloxy)ethyloxy); metal salts of salicylic acid, such as zinc salicylate and bis[4-(octyloxycarbonylamino)-2-hydroxybenzoic acid; hydroxysulfones, such as 4,4'-dihydroxydiphenylsulfone, 2,4'-dihydroxydiphenylsulfone, 4-hydroxy-4'-methyldiphenylsulfone, 4-hydroxy-4'-isopropoxydiphenylsulfone, 4-hydroxy-4'-benzyloxydiphenylsulfone, 4-hydroxy-4'-butoxydiphenylsulfone, 4,4'-dihydroxy-3,3'-

5-tert-butylphenyl)butane, and 1,1,3-tris(2-methyl-4-hydroxy-5-cyclohexylphenyl)butane have excellent effect against heat and humidity.

As the desensitizer, aliphatic higher alcohols, polyethylene glycol, guanidine derivatives and the like may be exemplified.

Examples for the antitack agent include stearic acid, zinc stearate, calcium stearate, carnauba wax, paraffin wax, and ester wax.

Examples for the photostabilizing agent include salicylic acid based ultraviolet radiation absorbents, such as phenyl salicylate, p-tert-butylphenyl salicylate and p-octylphenyl salicylate; benzophenone based ultraviolet radiation absorbents, such as 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-benzyloxybenzophenone, 2-hydroxy-4-octyloxybenzophenone, 2-hydroxy-4-dodecyloxybenzophenone, 2,2'-dihydroxy-4-methoxybenzophenone, 2,2'-dihydroxy-4,4'-dimethoxybenzophenone and 2-hydroxy-4-methoxy-5-sulfobenzophenone; benzotriazole based ultraviolet radiation absorbents, such as 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-5'-tert-butylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)benzotriazole, 2-(2'-hydroxy-3'-tert-butyl-5'-methylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3',5'-di-tert-amylphenyl)benzotriazole, 2-[2'-hydroxy-3'-(3",4",5",6"-tetrahydrophthalimidemethyl)-5'-tert-methylphenyl]benzotriazole, 2-(2'-hydroxy-5'-tert-octylphenyl)benzotriazole, 2-[2'-hydroxy-3',5'-bis(α,α -dimethylbenzyl)phenyl]-2H-benzotriazole, 2-(2'-hydroxy-3'-dodecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-undecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-undecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-tridodecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-tetradecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-pentadecyl-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-3'-hexadecyl-5'-methylphenyl)benzotriazole, 2-[2'-hydroxy-4'-(2"-ethylhexyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(2"-

ethylheptyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(2"-ethyloctyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(2"-propyloctyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(2"-propylheptyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(2"-propylhexyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-ethylhexyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-ethylheptyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-ethyloctyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-propyloctyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-propylheptyl)oxyphenyl]benzotriazole, 2-[2'-hydroxy-4'-(1"-propylhexyl)oxyphenyl]benzotriazole, and a condensate of polyethylene glycol and methyl-3-[3-tert-butyl-5-(2H-benzotriazole-2-yl)-4-hydroxyphenyl]propionate; cyanoacrylate based ultraviolet radiation absorbents, such as 2'-ethylhexyl-2-cyano-3,3-diphenylacrylate and ethyl-2-cyano-3,3-diphenylacrylate; and hindered amine based ultraviolet radiation absorbents, such as bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, succinic acid-bis(2,2,6,6-tetramethyl-4-piperidyl) ester and 2-(3,5-di-tert-butyl)malonate-bis(1,2,2,6,6-pentamethyl-4-piperidyl) ester.

Examples for the fluorescent dye include 4,4'-bis[2-anilino-4-(2-hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid disodium salt, 4,4'-bis[2-anilino-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid disodium salt, 4,4'-bis[2-methoxy-4-(2-hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid disodium salt, 4,4'-bis[2-methoxy-4-(2-hydroxypropyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid disodium salt, 4,4'-bis[2-m-sulfoanilino-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid disodium salt, 4-[2-p-sulfoanilino-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]-4'-[2-m-sulfoanilino-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid tetrasodium salt, 4,4'-bis[2-p-sulfoanilino-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid tetrasodium salt, 4,4'-bis[2-

(2,5-disulfoanilino)-4-phenoxyamino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid hexasodium salt, 4,4'-bis[2-(2,5-disulfoanilino)-4-(p-methoxycarbonylphenoxy)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid hexasodium salt, 4,4'-bis[2-(p-sulfophenoxy)-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid tetrasodium salt, 4,4'-bis[2-(2,5-disulfoanilino)-4-formalinylamino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid hexasodium salt and 4,4'-bis[2-(2,5-disulfoanilino)-4-bis(hydroxyethyl)amino-1,3,5-triazinyl-6-amino]stilbene-2,2'-disulfonic acid hexasodium salt.

The compounds of the present invention may be used for producing pressure-sensitive copying papers according to the same process for using a known image storing and stabilizing agent, a known developer and a known sensitizer. For example, a color forming dye prepared into a form of microcapsule is dispersed by using an appropriated dispersing agent according to a known procedure, and the resultant dispersion is coated onto a paper to obtain a sheet coated with a color forming dye. On the other hand, a dispersion of a developer is coated onto a paper to prepare a sheet coated with a developer. At that time, when the compound of the present invention is used as an image storing and stabilizing agent, the compound may be used by means of incorporating it into a dispersed solution of any component to be used for preparing a coupler sheet or a developer sheet. The both sheets prepared as described above are combined to prepare a pressure-sensitive copying paper. The pressure-sensitive copying paper may be an unit paper comprising an upper layer sheet holding a microcapsule layer enclosing an organic solvent solution of a color forming dye onto the underside face and an under sheet layer holding a developer (an acidic substance) onto the upper side face or self content paper coated with the microcapsule dispersion and a developer dispersion on the identical face of a support paper.

As the developer to be used for producing pressure-sensitive copying papers and the developer to be used in combination with

the compound of the present invention for the same application, known developers having been conventionally used may be used. Examples for such developers include inorganic acidic substance, such as acid clay, activated clay, apatite, bentonite, colloidal silica, aluminum silicate, magnesium silicate, zinc silicate, tin silicate, burned kaolin and talc; aliphatic carboxylic acid, such as oxalic acid, maleic acid, tartaric acid, citric acid, succinic acid and succinic acid; aromatic carboxylic acid, such as benzoic acid, p-tert-butylbenzoic acid, phthalic acid, gallic acid, salicylic acid, 3-isopropylsalicylic acid, 3-phenylsalicylic acid, 3-cyclohexylsalicylic acid, 3,5-di-tert-butylsalicylic acid, 3-methyl-5-benzylsalicylic acid, 3-phenyl-5-(2,2-dimethylbenzyl)salicylic acid, 3,5-di-(2-methylbenzyl)salicylic acid, and 2-hydroxy-1-benzyl-3-naphthoic acid; metal salts, such as zinc, magnesium, aluminum and titanium salts, of the above-mentioned aliphatic carboxylic acids; phenol resin based developers, such as p-phenylphenol-formalin resin and p-butylphenol-acetylene resin; and mixtures of said phenol resin based developer and a metal salt of said aromatic carboxylic acid.

Best Modes for Carrying Out the Invention

Now, the compounds of the present invention is further described in detail with referring the examples in the following. Note that the part indicated below in the examples denotes a part by weight.

Example 1

Synthesis of 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone
(Compound No. I-4)

10.0 g (79.4 mmol) of 4-mercaptophenol, 5.3 g (80.4 mmol) of potassium hydroxide and 100 mL of methanol were added under a cold temperature into a 200 mL flask with four inlets and attached with a stirrer and a thermometer. After confirming that potassium hydroxide added is completely dissolved, temperature inside the resultant solution was cooled down to 10°C, then 16.9

g (78.6 mmol) of 2'-hydroxyphenacyl bromide was added to the solution and stirred for 3 hours at a cold temperature. Following to the completion of the reaction, the solution was extracted with methyl isobutyl ketone, hereinafter referred to as MIBK, and MIBK was distilled out of the extract under reduced pressure. The obtained residue was subjected to recrystallization process with toluene to thereby obtain 19.0 g of 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone. The yield was 93% and the melting point thereof was in a range of 139 to 141°C.

Example 2

Synthesis of 2'-hydroxy-2-(4-hydroxyphenylsulfinyl) acetophenone (Compound No. I-5)

6.0 g (23.1 mmol) of 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone and 50 mL of acetic acid were added under a cold temperature into a 100 mL flask with four inlets and attached with a stirrer and a thermometer. To the resultant solution, 2.8 g (24.7 mmol) of 30% aqueous solution of hydrogen peroxide was added, and the solution was stirred for 12 hours at a cold temperature. Following to the completion of the reaction, 0.5 g of dimethyl sulfide was added into the solution, and then, the solution was extracted with MIBK. The MIBK layer was washed several times with water, and followed by washing with sodium hydrogencarbonate. The MIBK in the solution was distilled out under reduced pressure, and the resultant residue was subjected to recrystallization with ethyl acetate to obtain 4.5 g of 2'-hydroxy-2-(4-hydroxyphenylsulfinyl) acetophenone. The yield was 71% and the melting point of the compound was in a range of 166 to 167°C.

Example 3

Synthesis of 2'-hydroxy-2-(4-hydroxyphenylsulfonyl) acetophenone (Compound No. I-6)

6.0 g (23.1 mmol) of 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone and 50 mL of chloroform were added under a cold temperature into a 100 mL flask with four inlets and attached

with a stirrer and a thermometer. To the resultant solution, 11.2 g (48.5 mmol) of m-perchlorobenzoic acid (purity 75%) was added a few at a time under a cold temperature, and the solution was stirred for 4 hours. Following to the completion of the reaction, 0.5 g of dimethyl sulfide was added into the solution, and then, the solution was extracted with chloroform. The chloroform layer was washed with aqueous solution of sodium hydrogencarbonate. The chloroform in the solution was distilled out under reduced pressure, and the resultant residue was subjected to recrystallization with toluene to obtain 5.0 g of 2'-hydroxy-2-(4-hydroxyphenylsulfonyl)acetophenone. The yield was 74% and the melting point of the compound was in a range of 143 to 146°C.

Example 4

Synthesis of 4'-hydroxy-2-(4-hydroxyphenylthio)acetophenone (Compound No. I-106)

4'-hydroxy-2-(4-hydroxyphenylthio)acetophenone in an amount of 17.5 g was obtained by proceeding the same reaction and post-reaction as described in the Example 1, except that 2'-hydroxyphenacyl bromide is replaced by 4'-hydroxyphenacyl bromide. The yield was 86% and the melting point of the compound was in a range of 194 to 197°C.

Example 5

Synthesis of 4'-hydroxy-2-(4-hydroxyphenylsulfinyl) acetophenone (Compound No. I-107)

4'-hydroxy-2-(4-hydroxyphenylsulfinyl)acetophenone in an amount of 4.8 g was obtained according to the same process as described in the Example 2, except that 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone is replaced by 4'-hydroxy-2-(4-hydroxyphenylthio)acetophenone. The yield was 75% and the melting point of the compound was in a range of 167 to 169°C.

Example 6

Synthesis of 4'-hydroxy-2-(4-hydroxyphenylsulfonyl) acetophenone (Compound No. I-108)

4'-hydroxy-2-(4-hydroxyphenylsulfonyl)acetophenone in an amount of 5.4 g was obtained according to the same process as described in the Example 3, except that 2'-hydroxy-2-(4-hydroxyphenylthio)acetophenone is replaced by 4'-hydroxy-2-(4-hydroxyphenylthio)acetophenone. The yield was 80% and the melting point of the compound was in a range of 212 to 214°C.

Example 7

Synthesis of 2-(4-hydroxyphenylsulfinyl)acetoanilide (Compound No. II-1)

6.0 g (23.2 mmol) of 2-(4-hydroxyphenylthio)acetoanilide and 50 mL of acetic acid were added under a cold temperature into a 100 mL flask with four inlets and attached with a stirrer and a thermometer. To the resultant solution, 2.8 g (24.7 mmol) of 30% aqueous solution of hydrogen peroxide was added, and the resultant solution was stirred for 12 hours at a cold temperature. Following to the completion of the reaction, 0.5 g of dimethyl sulfide was added into the solution, and then, the solution was extracted with MIBK. The MIBK layer was washed several times with water, and followed by washing with sodium hydrogencarbonate. The MIBK in the solution was distilled out under reduced pressure, and the resultant residue was subjected to recrystallization with MIBK to obtain 5.9 g of 2-(4-hydroxyphenylsulfinyl)acetoanilide. The yield was 93% and the melting point of the compound was in a range of 208 to 210°C.

Example 8

Synthesis of 2-(4-hydroxyphenylsulfonyl)acetoanilide (Compound No. II-2)

6.0 g (23.2 mmol) of 2-(4-hydroxyphenylthio)acetoanilide and 50 mL of acetic acid were added under a cold temperature into a 100 mL flask with four inlets and attached with a stirrer and a thermometer. To the resultant solution, 5.6 g (49.4 mmol) of 30% aqueous solution of hydrogen peroxide was added, and the solution

was stirred for 4 hours at a cold temperature and consequently for 5 hours at 100°C. Following to the completion of the reaction, 0.5 g of dimethyl sulfide was added into the solution, and then, the solution was extracted with MIBK. The MIBK layer was washed several times with water, and followed by washing with sodium hydrogencarbonate. The MIBK in the solution was distilled out under reduced pressure, and the resultant residue was subjected to recrystallization with MIBK to obtain 5.8 g of 2-(4-hydroxyphenylsulfonyl) acetoanilide. The yield was 86% and the melting point of the compound was in a range of 188 to 189°C.

Example 9 (Preparation of Thermal Recording Papers)

Dye dispersion (A solution)

3-di-n-butylamino-6-methyl-7-anilino-fluoran	16 parts
10% aqueous solution of polyvinyl alcohol	84 parts

Developer dispersion (B solution)

4'-hydroxy-2-(4-hydroxyphenylsulfonyl)acetophenone (Compound No.I-108)	16 parts
10% aqueous solution of polyvinyl alcohol	84 parts

Filler dispersion (C solution)

Calcium carbonate	27.8 parts
10% aqueous solution of polyvinyl alcohol	26.2 parts
Water	71 parts

All components for each of A solution, B solution and C solution shown above were mixed and thoroughly grinded by using a sand grinder, respectively, to prepare each dispersed solutions of A to C. 1 part by weight of A solution, 2 parts by weight of B solution and 4 parts by weight of C solution were mixed to prepare a coating solution. The coating solution was coated onto a white paper by using a wire rod (No. 12) and then dried. The coated paper was then subjected to calendaring to prepare a thermal recording paper. (The amount of the coating solution based on the dry weight was approximately 5.5 g/m².)

The thermal recording material of the present invention was prepared according to the same process as described in Example 9, except that 4-hydroxy-4'-isopropoxydiphenylsulfone was used in place of the developer used in Example 9.

Comparative Example 2

The thermal recording material of the present invention was prepared according to the same process as described in Example 9, except that 2,4'-dihydroxydiphenylsulfone was used in place of the developer used in Example 9.

Comparative Example 3 (Compound disclosed in Jap. Pat. No. 2615073)

The thermal recording material of the present invention was prepared according to the same process as described in Example 9, except that 2-(4-hydroxyphenylsulfonyl)acetophenone was used in place of the developer used in Example 9.

Comparative Example 4 (Compound disclosed in Jap. Pat. Appln. KOKAI Publication No. 2-204091)

The thermal recording material of the present invention was prepared according to the same process as described in Example 9, except that 3',4'-dihydroxy-2-(4-hydroxyphenylsulfonyl)acetophenone was used in place of the developer used in Example 9.

Comparative Example 5 (Compound disclosed in Jap. Pat. Appln. KOKAI Publication No. 4-217657)

The thermal recording material of the present invention was prepared according to the same process as described in Example 9, except that 2-(4-hydroxyphenylthio)acetoanilide was used in place of the developer used in Example 9.

Test Example 1 (Comparison in Dynamic Sensitivity)

The thermal recording papers prepared in Examples 9 to 12 and Comparative Examples 1 to 4 were recorded under a condition

of 0.38 mJ and 0.50 mJ per dot by using Thermal Recording Paper Color Forming Testing Apparatus (manufactured by Okura Denki Co., Ltd., Type: TH-PMD), and the density of the images was measured by means of Macbeth densitometer, RD-514. The results are shown in Table 3.

Table 3

Evaluation Results of Dynamic Sensitivity		
	Quantity of Energy	
	0.38mj/dot	0.50mj/dot
Example 9	0.36	0.82
Example 10	0.42	0.90
Example 11	0.36	0.90
Example 12	0.33	0.94
Comparative Example 1	0.85	1.19
Comparative Example 2	0.57	1.15
Comparative Example 3	0.88	1.19
Comparative Example 4	0.21	0.47

*Figures indicated in the table denote Macbeth values.

Test Example 2 (Heat and Humidity Test)

Each of the thermal recording papers prepared in Examples 9 to 14 and Comparative Examples 1 to 5 were recorded according to the same procedures as described in Test Example 1. A heat and humidity test was conducted for the images being recorded to the saturated state in a thermohygrostat, Type: GL-42, manufactured by Futaba Science, under a temperature of 50°C and humidity of 80%. The density of the color formed images after 2 and 24 hours were measured. The results are shown in Table 4.

Test Example 3 (lightfast Test)

Each of the thermal recording papers prepared in Examples 9 to 14 and Comparative Examples 1 to 5 was recorded according to the same procedures as described in Test Example 1. The images were subjected to lightfast tests where a lightfast testing

apparatus (Ultraviolet Radiation Long Life Fade Meter, Type: FAL-5, manufactured by Suga Shikenki Co., Ltd.) is employed for the measurement. The densities of the tested images after 48 hours were measured. The results are shown in Table 4.

Table 4

(Evaluation Results on Backgrounds and Images)

	Background			Image				
	Original	Heat and Humidity		Original	Lightfastness			
		2hr	24hr		6hr	12hr	24hr	48hr
Example 9	0.05	0.05	0.05	1.13	1.10	1.06	1.03	0.95
					<98>	<94>	<91>	<85>
Example 10	0.06	0.07	0.07	1.12	1.14	1.09	1.06	0.90
					<102>	<97>	<94>	<80>
Example 11	0.05	0.05	0.05	1.17	1.11	0.98	0.91	0.74
					<95>	<83>	<78>	<63>
Example 12	0.07	0.06	0.06	1.32	1.29	1.26	1.33	1.35
					<98>	<95>	<101>	<102>
Example 13	0.06	0.06	0.06	1.23	1.20	1.21	1.13	1.07
					<98>	<98>	<92>	<87>
Example 14	0.05	0.05	0.05	0.58	0.65	0.57	0.55	0.48
					<112>	<98>	<95>	<83>
Comparative Example 1	0.08	0.07	0.07	1.26	1.08	0.60	0.29	0.14
					<86>	<48>	<23>	<11>
Comparative Example 2	0.10	0.10	0.10	1.25	1.19	1.08	0.96	0.76
					<96>	<87>	<76>	<61>
Comparative Example 3	0.11	0.16	0.18	1.22	1.23	1.11	1.00	0.59
					<101>	<91>	<82>	<48>
Comparative Example 4	0.09	0.09	0.09	1.06	1.03	0.96	0.82	0.72
					<97>	<91>	<77>	<68>
Comparative Example 5	0.04	0.04	0.05	1.25	1.23	1.14	1.07	0.89
					<98>	<91>	<85>	<72>

* Figures indicated in the table denote Macbeth values, and the figures in < > denote residual image ratio.

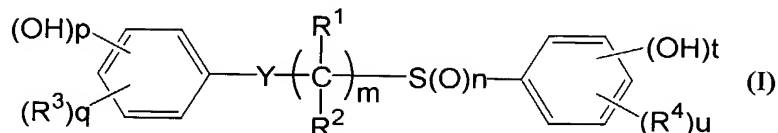
Advantageous Effect of the Invention

The recording material using the phenol compound of the present invention as a developer provides images with more improved storing and stabilizing property than images formed with

conventional recording materials. With the phenol compounds of the present invention, a recording material having excellent dynamic sensitivity and preservative properties of image and background can be obtained.

What is claimed is:

1. Phenol compounds represented by a general formula (I);



wherein R¹ and R² represent hydrogen or C1-C6 alkyl,

m represents an integer of 1 to 6,

n represents an integer of 0 to 2,

p and t represent an integer of 0 to 3, with proviso that p and t never be 0, concurrently,

R³ and R⁴ nitro, carboxyl, halogen, C1-C6 alkyl, C1-C6 alkoxy, C1-C6 alkoxy carbonyl, sulfamoyl, phenylsulfamoyl, C1-C6 alkylsulfamoyl, di(C1-C6 alkylsulfamoyl), carbamoyl, phenylcarbamoyl, C1-C6 alkylcarbamoyl or di(C1-C6 alkylcarbamoyl),

q and u represent an integer of 0 to 2,

R³ and R⁴ may be different to each other when q and u are 2,

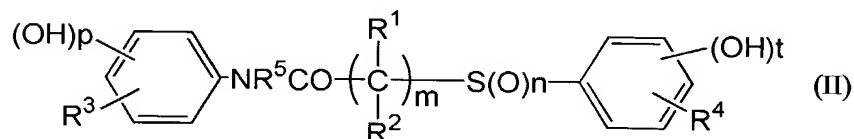
Y represents CO or NR⁵CO,

R⁵ represents hydrogen, C1-C6 alkyl, optionally-substituted phenyl or optionally-substituted benzyl,

with proviso that p is 1 when Y is CO, and

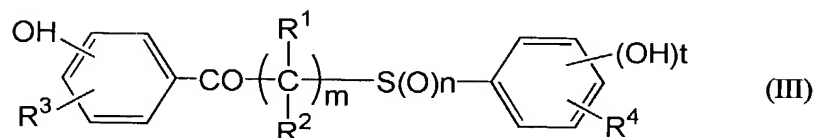
n is not 0 when p is 0 and Y is NR⁵CO.

2. Phenol compounds represented by a general formula (II);



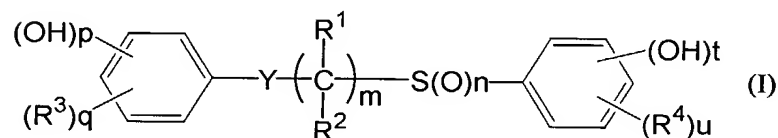
wherein R¹, R², R³, R⁴, R⁵, m, n, p and t are as defined above, with proviso that p and t may be 0.

3. Phenol compounds represented by a general formula (III);



wherein R^1 , R^2 , R^3 , R^4 , R^5 , m , n and t are as defined above, with proviso that t may be 0.

4. A recording material containing a color forming dye characterized in that the recording material comprises at least one of the phenol compounds represented by a general formula (I)



wherein R^1 and R^2 represent hydrogen or C1-C6 alkyl,

m represents an integer of 1 to 6,

n represents an integer of 0 to 2,

p and t represent an integer of 0 or 1, with proviso that p and t never be 0, concurrently,

R^3 and R^4 represent hydrogen, hydroxy, carboxyl, halogen, C1-C6 alkyl, C1-C6 alkoxy, C1-C6 alkoxy carbonyl, sulfamoyl, phenylsulfamoyl, C1-C6 alkylsulfamoyl, di(C1-C6 alkylsulfamoyl), carbamoyl, phenylcarbamoyl, C1-C6 alkylcarbamoyl or di(C1-C6 alkylcarbamoyl),

q and u represent an integer of 1 to 2, and

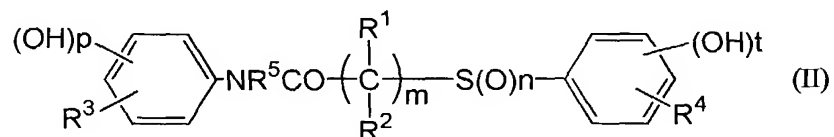
R^3 and R^4 may be different to each other when q and u are 2,

Y represents CO or NR^5CO ,

R^5 represents hydrogen, C1-C6 alkyl, optionally-substituted phenyl or optionally-substituted benzyl,

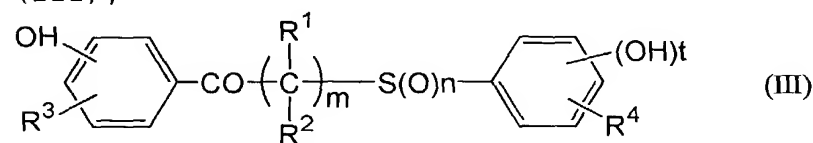
with proviso that p is 1 and R^3 is not hydroxy when Y is CO , and n and t are not 0 when p is 0 and Y is NR^5CO .

5. A recording material containing a color forming dye characterized in that the recording material comprises at least one of the phenol compounds represented by a general formula (II);



wherein R^1 , R^2 , R^3 , R^4 , R^5 , m , n , p and t are as defined above, with proviso that p and t may be 0.

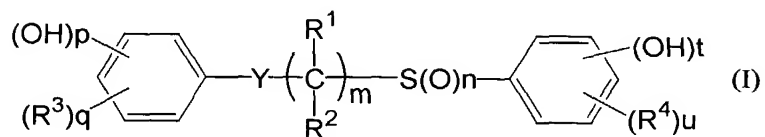
6. A recording material containing a color forming dye characterized in that the recording material comprises at least one of the phenol compounds represented by a general formula (III);



wherein R^1 , R^2 , R^3 , R^4 , m , n and t are as defined above, with proviso that t may be 0.

$$\begin{aligned} & \frac{\partial}{\partial t} \left(\frac{1}{\rho} \right) + \frac{\partial}{\partial x} \left(\frac{1}{\rho} u \right) + \frac{\partial}{\partial y} \left(\frac{1}{\rho} v \right) + \frac{\partial}{\partial z} \left(\frac{1}{\rho} w \right) = - \frac{1}{\rho^2} \frac{\partial \rho}{\partial t} \\ & \quad - \frac{1}{\rho^2} \frac{\partial \rho}{\partial x} u - \frac{1}{\rho^2} \frac{\partial \rho}{\partial y} v - \frac{1}{\rho^2} \frac{\partial \rho}{\partial z} w \end{aligned}$$

and recording materials characterized by containing one of them and exhibiting high light stability wherein R^1 and R^2 are each hydrogen or C_1 - C_6 alkyl; m is an integer of 1 to 6; n is an integer of 0 to 2; p and t are each an integer of 0 to 3 with the proviso that not both are simultaneously 0; R^3 and R^4 are each nitro, carboxyl, halogeno, C_1 - C_6 alkyl, or the like; q and u are each an integer of 0 to 2 with the proviso that when q or u is 2, R^3 s or R^4 s may be different from each other; and Y is CO or NR^5CO (wherein R^5 is hydrogen or the like), with the proviso that when Y is CO, p is 1 and that when p is 0 and Y is NR^5CO , n is not 0.



$$\begin{aligned} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \end{pmatrix} &= \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \end{pmatrix} = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \end{pmatrix} \quad (7) \end{aligned}$$

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

[illegible]**Full name of sole or first inventor**

INVENTOR			Country of of Citizenship
Given Name	Middle Initial	Family (Last) Name	
Tomoya		HIDAKA	Japan

RESIDENCE/Post Office Address	CITY	Country
2-4, Yusyudaihigashi, Ichihara-shi	Chiba	Japan

Tomoya Hidaka

Date: Mar. 5 2002

INVENTOR			Country of of Citizenship
Given Name	Middle Initial	Family (Last) Name	
<u>Shinichi</u>		<u>SATO</u>	Japan

RESIDENCE/Post Office Address	CITY	Country
1-4-17, Sakuradai, Ichihara-shi	<u>Chiba</u>	Japan

Shinichi Sato

Date: Mar, 5, 2002

INVENTOR			Country of of Citizenship
Given Name	Middle Initial	Family (Last) Name	
Tadashi		KAWAKAMI	Japan

RESIDENCE/Post Office Address	CITY	Country
2-4, Yusyudaihigashi, Ichihara-shi	<u>Chiba</u>	Japan

Tadashi Kanakami

Date: Mar. 5, 2002

☐ **Signature** for fourth and subsequent joint inventors. Number of pages added: ____.

☐ **Signature** by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. Number of pages added: ____.

☐ **Signature** for inventor who refuses to sign or cannot be reached by person authorized under 37 CFR 1.47. Number of pages added: ____.

☐ Added page for **signature** by one joint inventor on behalf of deceased inventor(s) where legal representative cannot be appointed in time. (37 CFR 1.47)

☐ Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (C-I-P) application. ☐ Number of pages added: ____.

**ADDED PAGE TO COMBINED DECLARATION
AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION
OR C-I-P APPLICATION**

**CLAIM FOR BENEFIT OF EARLIER U.S./PCT APPLICATION(S)
UNDER 35 U.S.C. SECTION 120**

I hereby claim the benefit, under Title 35, United States Code, Section 120, of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, Section 112, I acknowledge the duty to disclose information that occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application.

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. SECTION 120:				
U.S. APPLICATIONS		Status		
U.S. APPLICATIONS	U.S. FILING DATE	Patented	Pending	Abandoned
PCT APPLICATION DESIGNATING THE U.S.				
PCT APPLICATION NO.	PCT FILING DATE	U.S. APPLICATION NOS. ASSIGNED (IF ANY)		
PCT/JP00/06892	04 October 2000 (04.10.00)		X	

**ALL FOREIGN APPLICATION(S), IF ANY, FILED MORE THAN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION**

Country	Application No.	Filing Date
JAPAN	11/282577	14 Oct 1999 (04.10.99)
JAPAN	2000/37488	16 Feb 2000 (16.02.00)